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PROPERTIES OF COMPOSITE MATERIALS, REINFORCED BY TRICHITE FILAMENTS (WHISKERS)

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PROPERTIES OF COMPOSITE MATERIALS, REINFORCED BY TRICHITE FILAMENTS (WHISKERS) **/19

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A brief review is given over properties of composite materials, reinforced by whiskers formed of filaments of graphite, iron, silver, copper, quartz, zinc, cadmium, and aluminum oxide, with tabulation of their mechanical properties and stress-strain diagrams. The ultimate strength, at various stresses and strains, is calculated for a typical example of aluminum whiskers forming 50 vol.% in titanium-molybdenum alloys, certain steels, and columbium alloys. The use of a René 41 steel, reinforced with 50% of aluminum whiskers, will reduce the weight at a ratio of 4 to 1 in the construction of a pressurized enclosure, provided that the whiskers are oriented in accordance with the isotensoid. A composite material with 12 vol.% oriented aluminum whiskers will be about 10 times stronger than a nonreinforced light alloy. Author

In the DOC-AIR-ESPACE Review frequent mention has been ~~made~~ (see Bibliography at the end of this article) of the properties and the interest centered on very fine fibers, known under various designations such as "beards", "mustache", or by their English name "whiskers". However, their practical applica-

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** Numbers in the margin indicate pagination in the original foreign text.

tion has never been taken into serious consideration; therefore, this article, written by one of our foremost specialists, is to give information on the use of such fibers and indications as to what can be expected of them.

Summary

In this report, the following items are covered: strength of thin fibers; properties and manufacture of trichite filaments; mechanism and theoretical prediction of the resistance of composite materials reinforced by these filaments; preliminary results and advantages expected in various special cases.

The development of light-weight high-strength structures, which always has been an important objective in the aeronautical industry, has gained further interest in the field of space technology in view of the fact that any increase in mass will result in a proportionally much larger gain in range.

In the case of greatest interest for space vehicles, namely that of shells computed at the internal pressure (biaxial state of tension), the main mechanical characteristic is the ultimate tensile strength σ_t. Consequently, as in the case of tie rods, bracing wires, and - in a general manner - in all structural members that work in pure tension, the structure must be lighter in weight the higher the ratio of the above resistance to the density of the material $\frac{\sigma_t}{\rho}$.

Within the same trend of ideas, one could also make use of the ratio of σ_t to the specific weight $\frac{\sigma_t}{\pi}$, i.e., the specific length or the length of a fiber rupturing under its own weight. This concept is even more attractive since it permits an illustration of the essential result on which most interest in trichite filaments is centered, namely, that the characteristic length is greater the smaller the diameter of the fiber itself. Actually, this fully proved fact

is the reason for considering the use of materials composed of increasingly fine fibers, such as glass fibers and, more recently, so-called whiskers which latter are simple chains of single crystals which, in themselves, obey the general law, i.e., they have a specific length that is greater the smaller the cross section of the single crystal, with the theoretical atomic cohesion forming the limit.

We will not further discuss the properties of thin filaments in general, since this item has been exceptionally well covered in a previous report (Bibl.4), except that we wish to introduce the official scientific French designation in this article, namely, that of "trichite".

Obviously, because of their very nature, these thin filaments cannot be used like conventional materials. However, recently advantage was taken of their toughness and strength, by applying such filaments to the reinforcement of composite structures which, in this manner, can be made extremely light in weight, as demonstrated at the end of the present paper.

1. Properties of Trichite Filaments

Although the properties of whiskers, or crystallographic chains, have been demonstrated on iron, there are other bodies known whose filaments have similar characteristics: graphite, silver, copper, zinc, cadmium, and certain oxides such as of aluminum and zirconium, or other compounds or mixtures of /20
compounds such as quartz.

Table I gives a compilation of the properties of such filaments.

Several theories have been developed in order to explain the exceptional resistance of these filaments.

The most widespread theory consists in assuming that this high strength

TABLE I

MECHANICAL PROPERTIES OF VARIOUS TRICHITES

Materials	Modulus of Elasticity E	Stress at Rupture	Ratio $\frac{\sigma_t}{E}$
	(hb)	(hb)	
Graphite	[illegible]	62	[illegible]
Iron	20 000	1 300	[illegible]
Silver	7 700	168	1/46
Copper	12 500	300	1/42
Quartz	7 700	420	1/18
Zinc	10 500	225	1/47
Cadmium ...	7 000	90	1/77
Al_2O_3 ...	52 000	1 200	1/43

is due to the fact that the single crystals in question merely exhibit a tendency toward helicoidal dislocation and that the initiation of edge dislocations, which considerably reduce the strength, is practically nonexistent here which makes it possible to obtain extremely high maximum strain at rupture or ultimate strength (of the order of 1/10 to 1/40 of the elasticity modulus).

Other interesting mechanical properties of "whiskers" have been demonstrated. These include, in particular:

a) The modulus of elasticity of the filament may differ greatly from that of the standard material (varying between half and double the value);

b) Creep is entirely absent, at least up to relatively high temperatures even those close to the melting point.

The production of such filaments is still in its very beginnings. Until now, the most interesting filaments of this type have been produced only in relatively small dimensions, such as a length of 5 - 10 mm and a diameter vary-

ing between 1 and 10 microns. Quite a large volume of research still remains to be done in this particular field, first in the laboratory and thereafter on an industrial scale.

2. Manufacture of Whiskers

Four principal techniques can be considered here:

1) Chemical reduction of the halogenated metal compounds, by introducing hydrogen in the presence of a reducing metal (for example, zinc).

2) Electrolytic dissociation, starting with a fused salt of the material of which the filaments are to be produced.

3) Mechanical process, based on the observation that, if a mild metal is subjected to very high stresses, whiskers have a tendency to develop in the core of the material (in fact, this process was originally responsible for drawing attention to the presence of such filaments).

4) Vapor deposition. Certain types of such filaments (Al_2O_3) can be obtained by depositing a supersaturated metal vapor on a surface.

Up to now, only chains of crystals of sufficiently small dimensions have been obtained, but it can be hoped that certain of the processes mentioned above will finally permit the production of chains which are sufficiently long and much finer.

It should be mentioned also that the obtained filaments may be of two different categories (although one cannot completely control the selection). In one of these categories, the chain formation develops parallel to the crystallographic axis whereas, in the other category, it develops perpendicular to this axis.

3. Mechanism of Strength of Composite Materials, Reinforced by Fibers

Let us now consider the case of a composite material, reinforced by filaments of trichite. For determining the optimum constitution of such a material, it is obviously necessary to understand the mechanism of reinforcement itself.

Let us assume first that we have to do with a uniaxial state of stress and that the filaments run parallel to the principal stress.

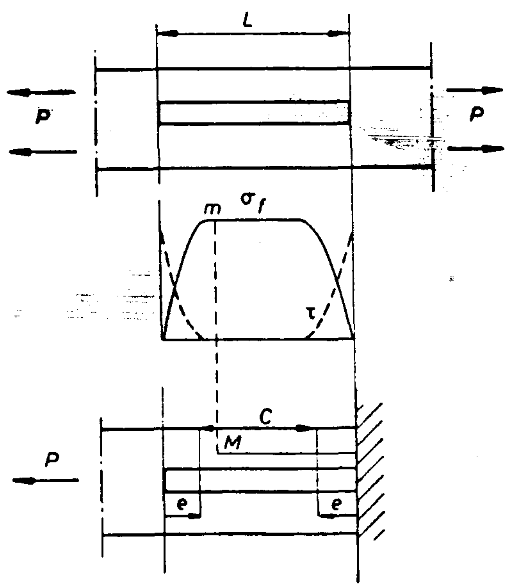

Fig.1 Mechanism of Transfer of the Load from Matrix to Filament

The fundamental hypothesis stipulates, for studying these composite /21 structures, that the filaments themselves absorb the largest portion of the applied load.

Obviously, the filaments also are able to reinforce the matrix itself to a certain degree, by opposing the motion of dislocations and by retarding the propagation of cracks. However, their main role rests in their strength itself.

Under these conditions and using our hypotheses, as soon as a tensile load

is applied to the composite material, one can assume that this load is transferred from the matrix to the filament over the intermediary of the shearing forces that manifest themselves on the interface between matrix and filament.

Figure 1 gives a schematic view of the above-described mechanism for the case of a unique filament, included in a metallic material. The diagram indicates that the tensile stress in the filament σ_f shows a flattening in the central portion c whereas the shearing stresses τ which transfer the load from the matrix to the filament, conversely, show a maximum at the extremities of the fiber where the matrix normally undergoes the greatest deformation.

In fact, it is logical to assume that the deformation of the matrix remains within the elastic domain, at the level of the central portion of the filament c, but reaches the plastic domain at the height of the two extremities e of this filament.

Based on these hypotheses, N.F.Dow demonstrated theoretically that the magnitude of shearing and tensile stresses, at the abscissa z, depends on

1) ratio of length of the filament to diameter of the filament L/d;
2) number of filaments per unit section ν;
3) applied load P;
4) physical properties of the filament, of the matrix, and of the interface junction line.

Under these conditions, the state of stress at the abscissa z is controlled by the following relations where m and f, respectively, characterize the matrix and the filaments:

$$P = P_m + P_f$$
$$P_{eff} = P_m - P_f\left(\frac{E_m S_m}{E_f S_f}\right), \quad (1)$$

$$\tau = \frac{\lambda}{4} \frac{P_{eff}}{S_m \frac{E_m}{E_f} + S_f} \frac{E_m S_m}{E_f S_f} \frac{\left(\frac{\lambda z}{d_f}\right)}{\cosh\left(\frac{\lambda z}{d_f}\right)}, \tag{2}$$

with

$$\lambda = 2\left[\frac{2\sqrt{2} + \left(\frac{G_f}{E_f}\right)\left[1 + \frac{S_f}{S_m}\frac{E_m}{E_f}\right]}{\sqrt{2} - 1 + \frac{G_f}{G_m}\left[\left(2 + \frac{S_m}{S_f}\right)^{1/2} - \sqrt{2}\right]}\right]^{1/2}, \tag{3}$$

$$\Big\} \quad \sigma_m = \frac{P_m}{S_m} \qquad \sigma_f = \frac{P_f}{S_f}. \tag{4}$$

The numerical results given in Figs.2, 3, and 4 refer to aluminum filaments in an aluminum matrix. It has been assumed here that the filament always is within the elastic domain, having an elasticity modulus of 42,000 hb and a breaking strength of 420 hb, as well as that the ratio of the cross sections of the matrix and the filaments is 100 $\left(\frac{S_m}{S_f} = 100\right)$ where it is understood that S_m exactly denotes the area of the portion of the matrix section which is actually influenced by the presence of the fiber, i.e., an area which must be experimentally determined.

A study of the diagrams shows that, as long as the matrix remains in the elastic domain, the load taken up by the filament remains relatively low in view of the fact that the elongation is too limited to produce an appreciable resilience. Conversely, as soon as the deformation of the matrix reaches the plastic domain, the participation of the filament in the resistance becomes much more extensive. However, the length necessary for ensuring a satis- /22
factory transfer of the load from the matrix to the fiber also increases, which means that the filament should have a length of 15 - 20 diameters.

This length for transfer can be reduced by increasing the Young's modulus of the matrix, for example, by increasing the density of the filaments. Dow demonstrated that, in the limiting case in which $E_m = E_f$, this transfer length

is reduced to 5 diameters.

The shearing stresses at the two extremities of the filament are never very high so long as the deformation of the matrix remains below the elastic

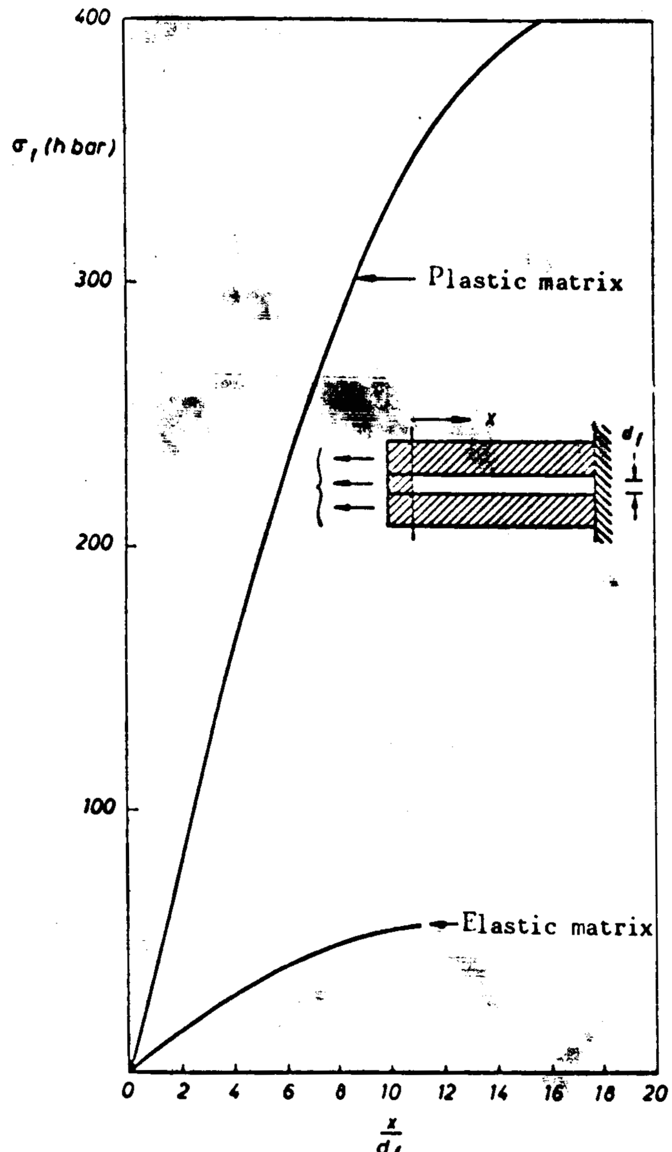

Fig.2 Tensile Stress in a Filament of Al_2O_3

$$\left(\frac{S_m}{S_f} = 100\right)$$

limit whereas, beyond this point, they assume appreciable magnitudes. Such stresses may also be quite high locally in the case in which the transfer sur-

faces have been considerably reduced, as was the case discussed in the preceding Section.

This indicates that the most difficult problem to be solved in order that these filaments can properly fill their role of reinforcement is the problem of

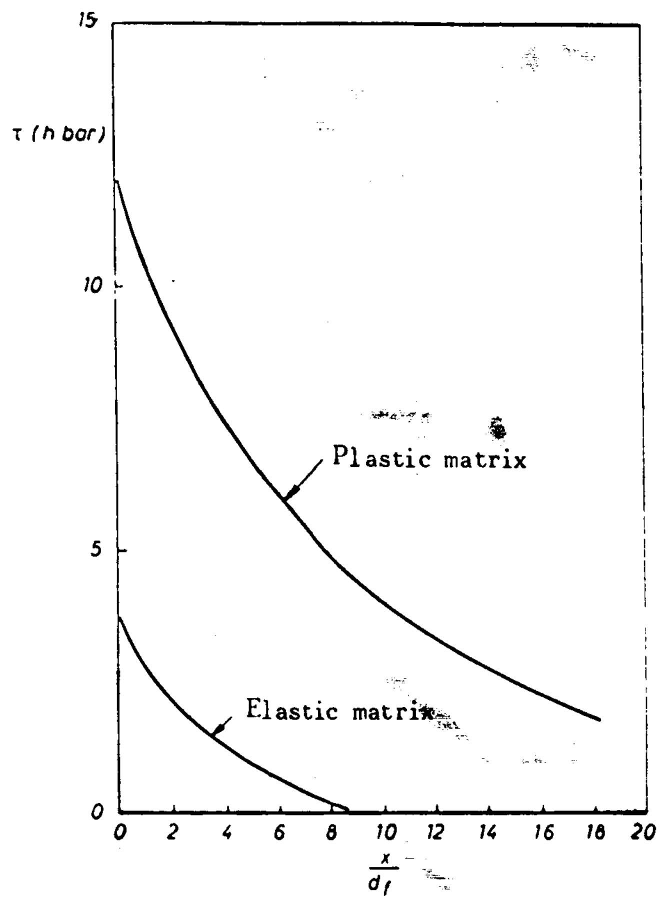

Fig.3 Shearing at the Interface. Matrix Al; filament Al_2O_3

passage of the shear to the extremities of the strands, at the level of the interfaces between metal and filament. This can be obtained by producing a joint that is simultaneously strong and flexible.

In addition to this problem of bonding, the selection of the material of the matrix and that of the relative length of the filaments (L/d) must be speci- /23

fically studied in order to obtain composite materials whose qualities correspond to the results predicted by the theory.

4. Theoretical Prediction of Mechanical Properties of Composite Materials

No general theory is in existence for predicting the properties of these composite materials, mainly due to the fact that there are no experimental data

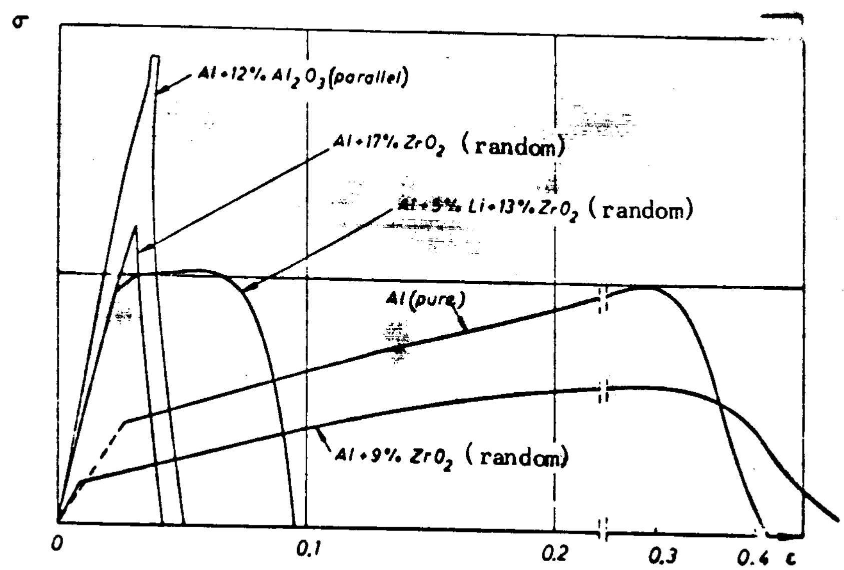

Fig.4 Stress-Strain Diagram for Matrices with an Aluminum Base

available which would yield the necessary approximations for the development of such a theory.

However, a very simple method is based on the hypothesis that the matrix and the filaments have the same elongation and both obey Hooke's law (elastic domain). Since the trichites are more rigid than the matrix, they are subject to a much higher rate of stress at the same assumed strain.

Under these conditions and knowing the stress-strain relation of each of the two components (matrix and filaments), the share of load of each can be

calculated as a function of the total deformation. From this, the so-called equivalent stress, given by the following formula, can be calculated:

$$\sigma_e = k_f S \sigma_f + k_m \sigma_m$$

where k_f and k_m, respectively, represent the fractions of the total sections occupied by the filaments on the one hand and by the matrix on the other hand:

$$k_f + k_m = 1.$$

Such a reasoning, however, can be valid only under the following conditions:

1) It is assumed that the ratio of length to diameter of the filament is of the order of 1000, so that one can be certain that the filament actually absorbs the maximum possible load (one actually knows how to produce filaments that satisfy this condition).

2) In determining the unit strength of the filaments, a corrective factor for dispersion is taken into consideration. The total strength of a bundle of fibers, actually, is less than the arithmetic mean of the strength of the individual fibers. The corrective factor S, inferior to 1, will restore the difference.

3) It is assumed that all the filaments are mutually parallel and also parallel to the direction of the principal stress. If this is not the case, an orientation factor θ must be introduced, varying from 1 at perfect orientation to 0.18 at purely random orientation.

4) It is recalled that, at high temperatures, the strength of the filament decreases with time.

Under these conditions, the equivalent stress in the composite material can be written as follows:

$$\sigma_a = k_f \, S \, 0 \, \bar{\sigma}_f + k_m \, \sigma_m,$$

an expression which clearly shows that the effect of reinforcement of the filaments in a composite material may be extremely reduced unless special care is taken in producing these filaments.

5. Preliminary Experimental Results

The most direct technique of manufacturing a composite material, reinforced by trichite filaments, consists in introducing, in vacuo, into the molten metal bath a cluster of such filaments previously coated with a special product that ensures satisfactory behavior of the interfaces. /24

The General Electric Company recently published several experimental results obtained with composite materials manufactured by this process, starting from a matrix of light alloys and filaments of zirconium or aluminum.

In a first case (9% by volume of ZrO_2), the properties of the composite material were less satisfactory than those of the pure material (random arrangement of the reinforcements).

However, if a lithium alloy (5 vol.%) is added, which improves the bonding of the interfaces, more satisfactory properties as well as a reduction in the elongation at rupture will be obtained.

The composite material with 17 vol.% of zirconium, with random orientation, is even superior from the viewpoint of rupture, showing only negligible elongation (no plastic domain).

A composite with 12 vol.% of aluminum, with non-random orientation, is about ten times superior to a nonreinforced light alloy.

6. Expected Gain in Various Cases

The filament of the type considered here is an aluminum filament (Al_2O_3), incorporated at 50 vol.% in the following materials:

a titanium-molybdenum alloy;

a steel of the René-41 type;

a columbium alloy.

It has been assumed here that the filaments were well aligned and parallel to the direction of the applied force and that the coefficient S, taking care

TABLE II

RESULTS EXPECTED FOR VARIOUS TYPES OF COMPOSITE MATERIALS, REINFORCED BY ALUMINUM WHISKERS (50 vol.%)

Base Material	$\frac{\sigma}{\rho}$ natural	$\frac{\sigma}{\rho}$ reinf.	Gain Factor
Light Al[illegible]AU 4G 1 ...	16		
Steel type PH 15-7. Mo ...	20		
Titanium Alloy	12.5	55	4.4
Steel type René 41	21	67	3.2
Columbium Alloy ...	13.4	59	4.4

of the standard deviation, was equal to 0.9 and that operation was in the vicinity of the ambient temperature.

Under these conditions, the results shown in Table II are obtained.

These data show that if, within a pressurized enclosure, the light alloy is replaced by a composite material of René 41 steel reinforced with 50% of aluminum filaments, the weight will be reduced at a proportion of 100 to 24 kg

or, roughly, at a ratio of 4 to 1, if the whiskers are oriented in accordance with the isotensoid.

BIBLIOGRAPHY

1. Hoffman, G.A.: The "Whiskers" (Les "whiskers"). Journal of Metals, September, 1958.
2. Sutton, W.M.: Development of Composite Structural Materials for Space Vehicles Applications. A.R.S. Journal, March 1962.
3. Bushong, R.M.: Graphite and its Applications in Space (Le graphite et ses applications spatiales). Report by O.Brodin, Doc-Air-Espace, No.84, p.70.
4. Wagner, H.J.: Reinforcement of Refractory Structural Materials by Filaments and Whiskers (Renforcement des matériaux structuraux réfractaires par des filaments et des barbes). Battelle Technical Review, December 1963, Report by P.Lefèvre, Doc-Air-Espace, No.87, p.75.

Readers interested in the problems discussed here are referred to the list of references at the end of the article by Wagner (Bibl.4).